

MATERIALS SCIENCE AND ENGINEERING

SEMINAR

“Thin Film Mixed Conducting Cathodes for Intermediate Temperature Solid Oxide Fuel Cells”

by

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ABSTRACT

Research efforts in the SOFC area have been directed towards systems operating at reduced temperature (500-700°C) while achieving high electrical performance. Since the cell electrical performance is limited by cathode processes, different cathode systems have been examined to decrease the cell electrical resistance. Mixed ionic-electronic conductors with high catalytic activity for oxygen reduction, such as $\text{La}_{0.6}\text{Sr}_{0.4}\text{Co}_{0.2}\text{Fe}_{0.8}\text{O}_{3-\delta}$ (LSCF), exhibit a low cathode polarization resistance. For enhanced cathode performance at intermediate temperature, composite structures of LSCF and ionic conductors, such as $\text{Ce}_{0.8}\text{Gd}_{0.2}\text{O}_{2-\delta}$ (CGO) can be employed. Reduction of cell operating temperature is also achieved by reducing the size scale of SOFCs. The availability of thin film deposition techniques and micro fabrication technology has sparked interest in micro-scale SOFCs consisting of electrode and electrolyte layers with submicron thicknesses.

This work was focused on the processing and characterization of thin (200-300 nm) film LSCF-CGO composite cathodes. The films were deposited on YSZ substrates to simulate the electrolyte/cathode interface in micro-scale SOFCs. Spray pyrolysis, which involves atomizing a precursor solution onto a heated substrate, was employed. A solution of nitrate salts dissolved in a mixture of ethanol and diethylene glycol was sprayed on a sintered YSZ laminate held at 235°C, with an air pressure of 0.14 MPa, a solution flow rate of 0.34 mL/min and with the airbrush positioned 20 cm above the substrate. The as-deposited films were amorphous and were heated to 500-900°C for crystallization to occur. The film topography displayed characteristic features of spray pyrolyzed films. In this study, a fractal dimension of 2.65 was estimated through roughness measurements at different scan sizes of 10-100 μm . A single precursor solution was employed to process the LSCF-CGO films. Phase segregation was observed by x-ray diffraction upon annealing above 600°C. Grain growth in the LSCF-CGO films was followed at 700-900°C and occurred to lesser extent than in LSCF films due to the CGO phase. The polarization resistance of the films with 0, 41, 66 and 100 vol% CGO fraction and annealing temperatures ranging from 500 to 900°C was studied by impedance spectroscopy at 350- 600°C. Several models proposed for mixed conducting cathodes were examined the fit the response of the thin films using complex non-linear least squares analysis. A model developed by Adler, Lane and Steele, which includes contribution from oxygen diffusion, surface exchange processes and microstructural parameters, was selected based on fit quality and confidence interval of parameter estimates. The magnitude and activation energy of the polarization resistance decreased with CGO addition, which was consistent with the properties of CGO. The resistances of LSCF and 59LSCF-41CGO films annealed at 700°C were 17 and 9 $\text{ohm}\cdot\text{cm}^2$ at 500°C. The variation of polarization resistance with increasing annealing temperature displayed a “v” shaped curve, which was related to nucleation and grain growth during film crystallization at 500-900°C. The resistance of 59LSCF-41CGO films annealed at 500, 700 and 900°C were 290, 106 and 256 $\text{ohm}\cdot\text{cm}^2$ at 400°C. The respective activation energies estimated over the 350-600°C operating temperature range were 1.45, 1.11 and 1.04 eV. Further characterization of phase segregation and crystallization with in situ x-ray diffraction and differential scanning calorimetry are recommended to compare the crystallization rates of the individual phases to those of the composite films. Measurements of the electrical properties in various oxygen atmospheres are also recommended to validate the physicochemical model employed to interpret the impedance results.

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